Isomorphic Phase Transformation in Shocked Cerium Using Molecular Dynamics

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Ce undergoes a significant (~16%) volume collapse associated with an isomorphic fcc-fcc phase transformation when subjected to compressive loading. We here present a new EAM potential for Cerium that models two minima for the two fcc phases. We show results from its use in the MD simulations of Ce samples subjected to shocks with pressures ranging from 0.5 to 25 GPa. We observed a split wave structure, with an elastic precursor followed by a plastic wave. The plastic wave causes the expected fcc-fcc phase transformation. Comparisons to experiments and MD simulations on Cesium indicate that three waves could be observed. The construction of the EAM potential may be the source of the difference.

Cerium (Ce) has an atypical phase diagram, presenting, among other things, an isomorphic phase transition between two face centered cubic (fcc) structures a and g. The g-to-a phase transition is believed to be caused by the transfer of valence electrons from the 4f state to the 5d state and induces a volume collapse of ~16% [1,2]. Shock loading experiments show a two- or three-wave profile, depending on the shock pressure [2,3]. The three-wave profile consists of an elastic precursor, a plastic wave in the g phase, and the g-to-a phase transformation. In the experiments leading to a two-wave profile, the elastic precursor is overdriven by the plastic waves.

We used a modified Voter-Chen [4] Embedded Atom Method (MVC-EAM) potential to fit the properties of Ce. This potential exhibits two minima for each of the FCC phases of Ce. An artificial energy barrier was introduced between the two minima so that each phase is stable. A few properties of this potential were calculated, including melting temperature, surface energies, and stable and unstable stacking fault energies.

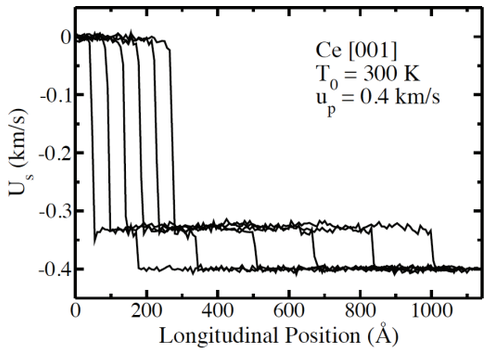

Fig. 1. Velocity profiles, spaced 4 ps apart, of Ce shocked in the [001] direction with a particle velocity of 0.4 km/s. These velocity profiles show a two-wave structure: an elastic precursor and a phase transformation accompanied by plasticity.

We use the LANL SPaSM (Scalable Parallel Short-range Molecular Dynamics) code, with 40 processors per simulation run. Single crystals of the g phase with two different crystallographic orientations are studied: <001> or <111> along the shock direction. The samples are about 13 nm × 13 nm × 130 nm and are composed of around 0.5 million atoms. Periodic boundary conditions are used in the lateral directions. The samples are studied at 10K and 300K. The shock wave is produced by launching the sample into a "momentum mirror" reflecting boundary with a specified velocity up.

A typical velocity profile is shown in Fig. 1. Two shock waves can be observed in this example. The first wave is an elastic precursor. In order to determine the nature of the second wave, a shocked sample is represented with its angle distribution analysis, as well as the radial distribution function (RDF) of the sample before and after the shocks (Fig. 2). The angle distribution analysis shows that stacking faults have been left behind partial dislocations after the second shock wave. Plasticity is thus sustained after the second shock, and at the same time, the lattice parameter after the second shock has decreased below the value of the lattice parameter of the a phase (4.85 Å). The phase transformation thus occurs before any plasticity has been sustained in the original g phase. This can be explained by looking at the energies required to initiate the phase transformation and the energy required to nucleate a stacking fault in the g phase (unstable stacking fault energy). The energy barrier between the two phases was chosen to be 0.142 eV/atom. In comparison, the unstable stacking fault energy in the g phase is 773 mJ/m^2, which corresponds to 0.245 eV/atom for the two layers of atoms on each side of the stacking fault. It is thus understandable that the phase transformation will occur before any plasticity can be sustained in the g phase.

We show in Fig. 3 the Us-up Hugoniots for the two loading directions and the two temperatures, along with a fit to experiments [5]. We first observe that our simulations reasonably agree with the experimental data, especially at low up. The orientation of the sample and the initial temperature have very little effect on the simulations, and especially on

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the phase transformation. The time and length scale of our simulations vary greatly with those of the experiments and might account for the differences. In addition, our samples are in a perfect lattice at the beginning of the simulation and are single crystalline. Other potential formalisms, in particular Ackland's two-band model [6], are also being investigated in order to study this electronically driven transition.

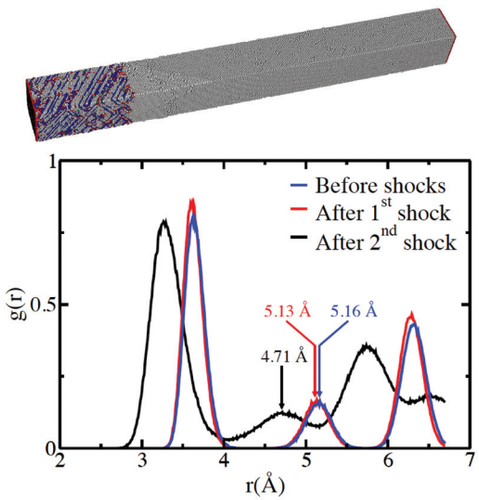

Fig. 2. (Top) Angle distribution analysis representation on a sample shocked in the [001] direction at 300K with up = 0.4 km/s. Grey: fcc, Blue: hexagonal close-packed (HCP), Red: unknown. (Bottom) RDF on slices after each shock. These figures show that plasticity occurs after the phase transformation.

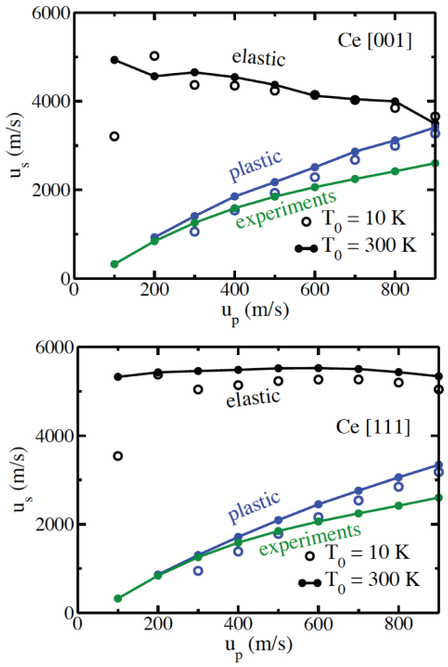

Fig. 3. Us-up Hugoniots of Ce for two different orientations and initial temperatures. The curves labeled "experiments" correspond to a fit to experiments from [5].

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Funding Acknowledgments
DOE, NNSA, Advanced Simulation and Computing (ASC) Program